Localized Patterns in Experimental Reaction-Diffusion Systems

Irving Epstein

Brandeis University

http://hopf.chem.brandeis.edu

With thanks to: Milos Dolnik, Marcin Leda, Vladimir Vanag, Lingfa Yang, Anatol Zhabotinsky

N$F

Localized Patterns

- Definition, types of patterns
- Patterns induced by global feedback and periodic illumination
- Spontaneous patterns in microemulsions
- Models and simulations (if time permits)
- Open questions

What is a localized pattern?

Vanag and Epstein,

Localized Patterns in Reaction-Diffusion Systems,

Chaos 17, 037110 (2007):

"A localized pattern consists of one or more regions in one state, typically characterized by a set of concentrations, temperature and/or other variables, surrounded by a region in a qualitatively different state. Such patterns may be stationary or oscillatory, static or moving."

Where do localized patterns occur?

- Fluid convection
- Semiconductors
- Gas discharges
- Granular materials
- Epilepsy
- Blood clots
- Reaction-diffusion systems

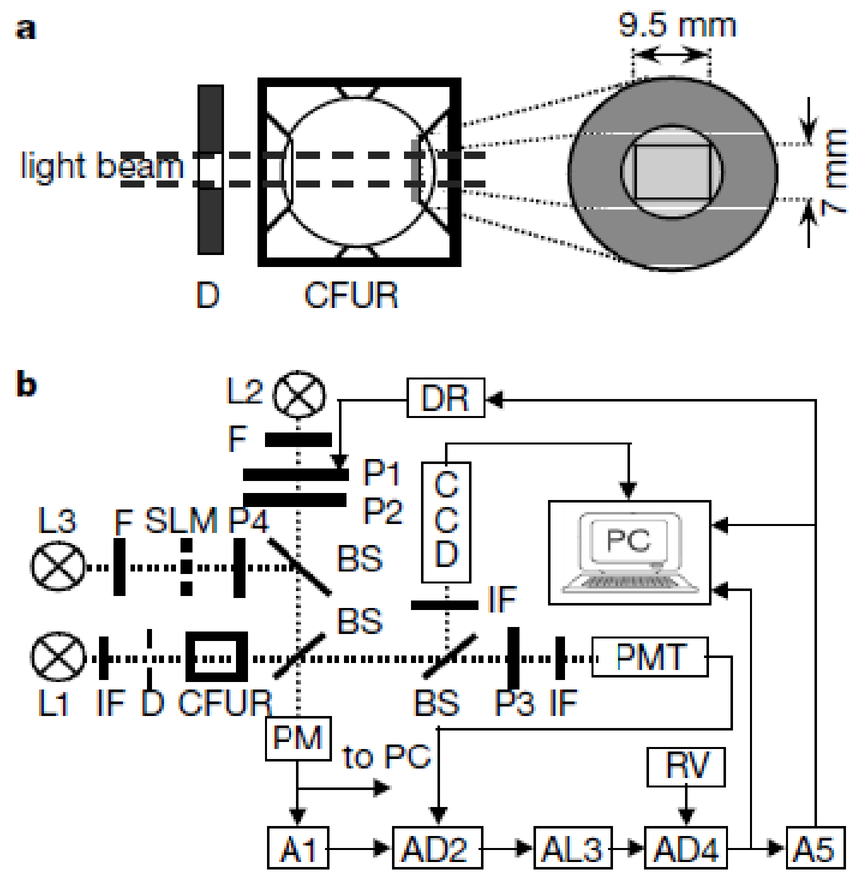
Types of localized patterns

- Stationary spots
- Oscillatory spots (oscillons)
- Breathing spots
- Oscillatory clusters
- Wave segments
- Moving spots
- Localized waves (solitons)

Localized patterns in the photosensitive BZ reaction

- Belousov-Zhabotinsky (BZ) oscillating chemical reaction (bromate, malonic acid, catalyst in acidic solution)
- Gives time-periodic oscillation, traveling spiral and target patterns (extended)
- With $Ru(biby)_3$ catalyst, reaction is photosensitive, light suppresses oscillation
- Can be run in a gel to suppress convection

Experimental setup



V.K. Vanag, L. Yang, M. Dolnik, A.M. Zhabotinsky IRE, Nature 406, 389 (2000).

Global feedback experiments

- To generate global negative feedback, intensity of illumination is varied as

$$I = I_{max} \sin^2[g(Z_{av} - Z_t)]$$

g = feedback coefficient (bifurc. param.)

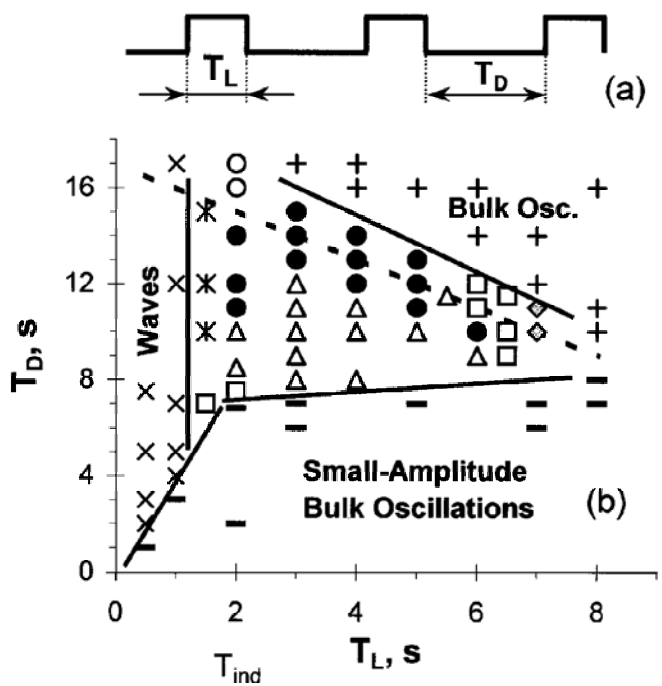
I_{max} = maximum intensity

Z_{av} = spatially averaged $[Ru(bipy)_3^{3+}]$

Z_t = target concentration (near steady state)

Periodic oscillation experiments

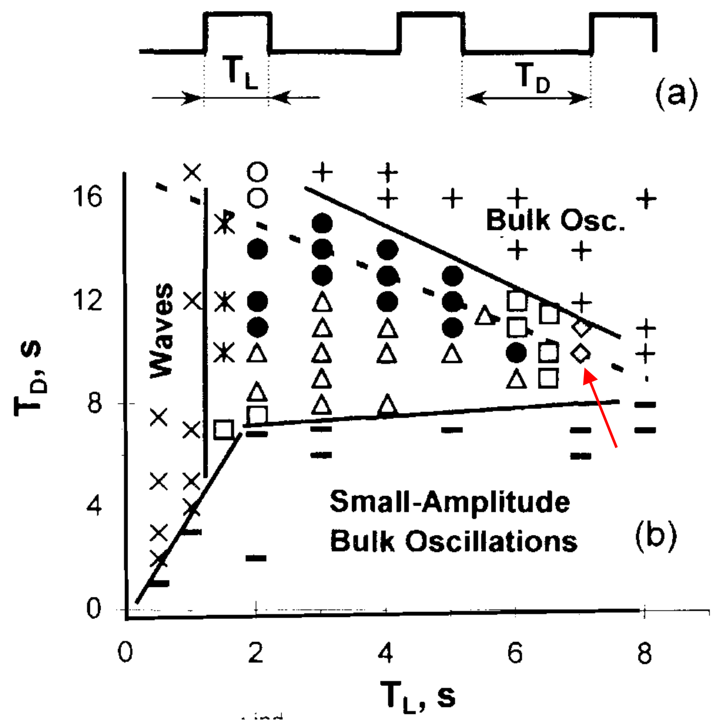
- Illumination is applied as square pulses with length of dark and light periods varied independently (free oscillator period = 17s)

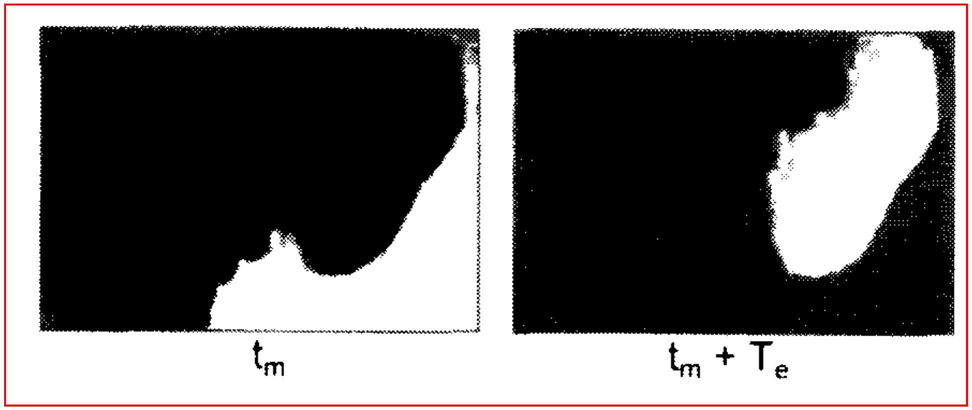


Experimental results

- As the bifurcation parameter (feedback coefficient or length of light pulse) is increased, observe transitions from bulk oscillation to clusters to steady state
- Cluster patterns consist of domains in which nearly all the elements oscillate with the same amplitude and phase. They arise in models of coupled neurons. Typically one finds 2 or 3 distinct phases (2- or 3-phase cluster patterns).

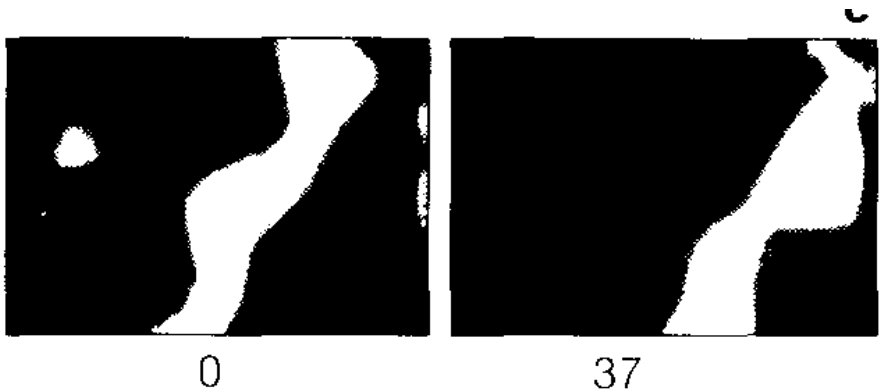
Periodic Forcing Experimental Results

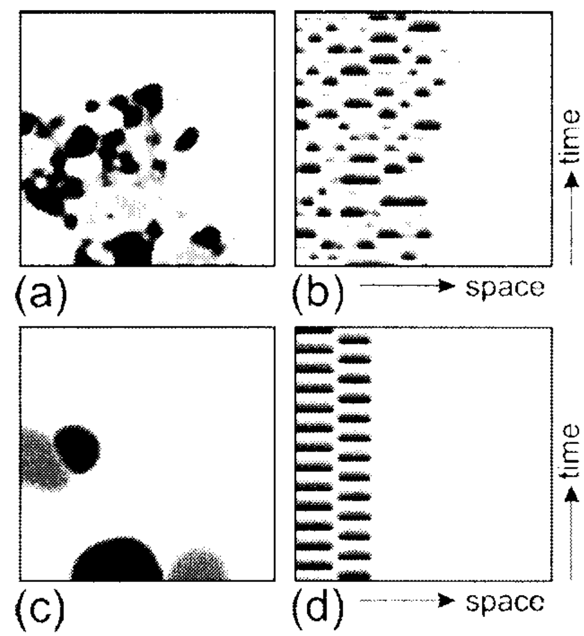




V. K. Vanag, A. M. Zhabotinsky and IRE, Phys. Rev. Lett. 86, 552 (2001)

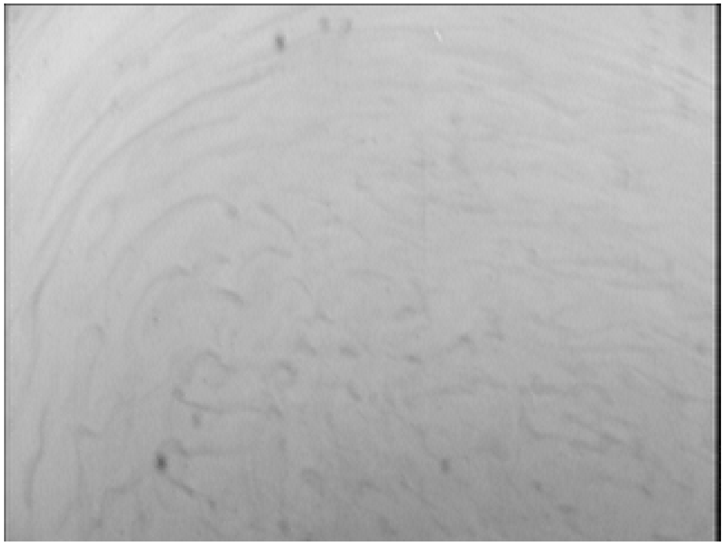
Global Feedback
Experimental Results

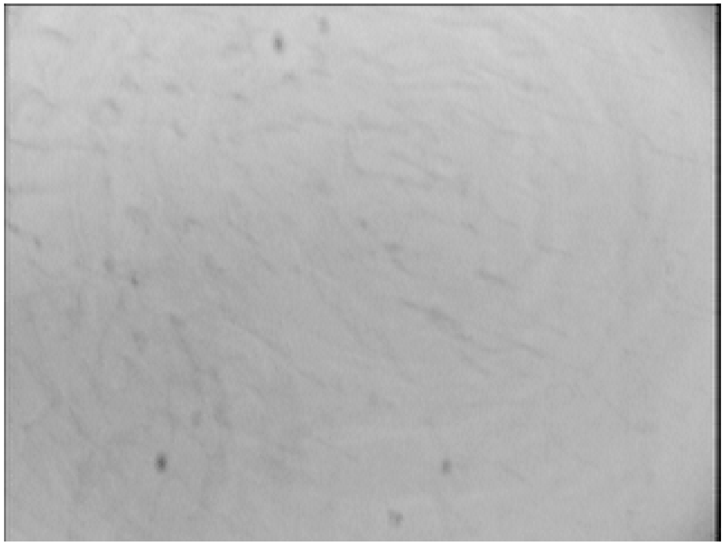




V.K. Vanag, A.M. Zhabotinsky and IRE, J. Phys. Chem. A 104, 11566 (2000).

Global Feedback
Experimental Results

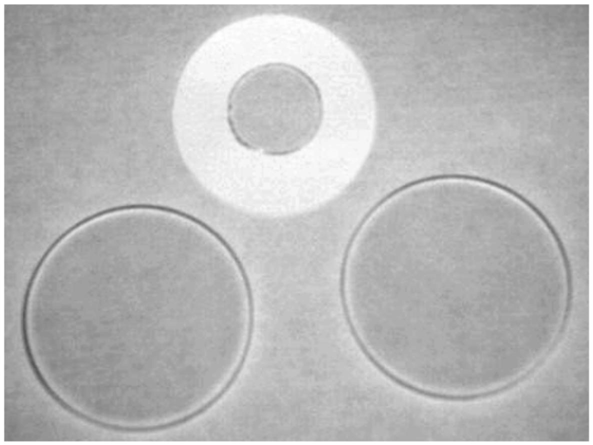


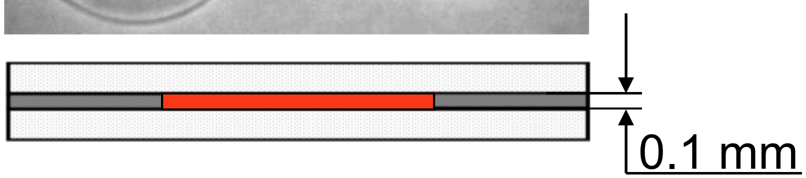


Microemulsion experiments – The BZ-AOT system

- AOT = aerosol OT (sodium bis(2-ethylhexyl) sulfosuccinate)) – surfactant in a water-in-oil (reverse) microemulsion
- Reaction takes place in 5-10 nm diameter water droplets
- Fast time scale – diffusion of nonpolar intermediates through the oil (octane)
- Slow time scale – collision and exchange of polar species between water droplets
- Cross-diffusion also occurs
- Rich variety of spatio-temporal patterns

Experimental Setup

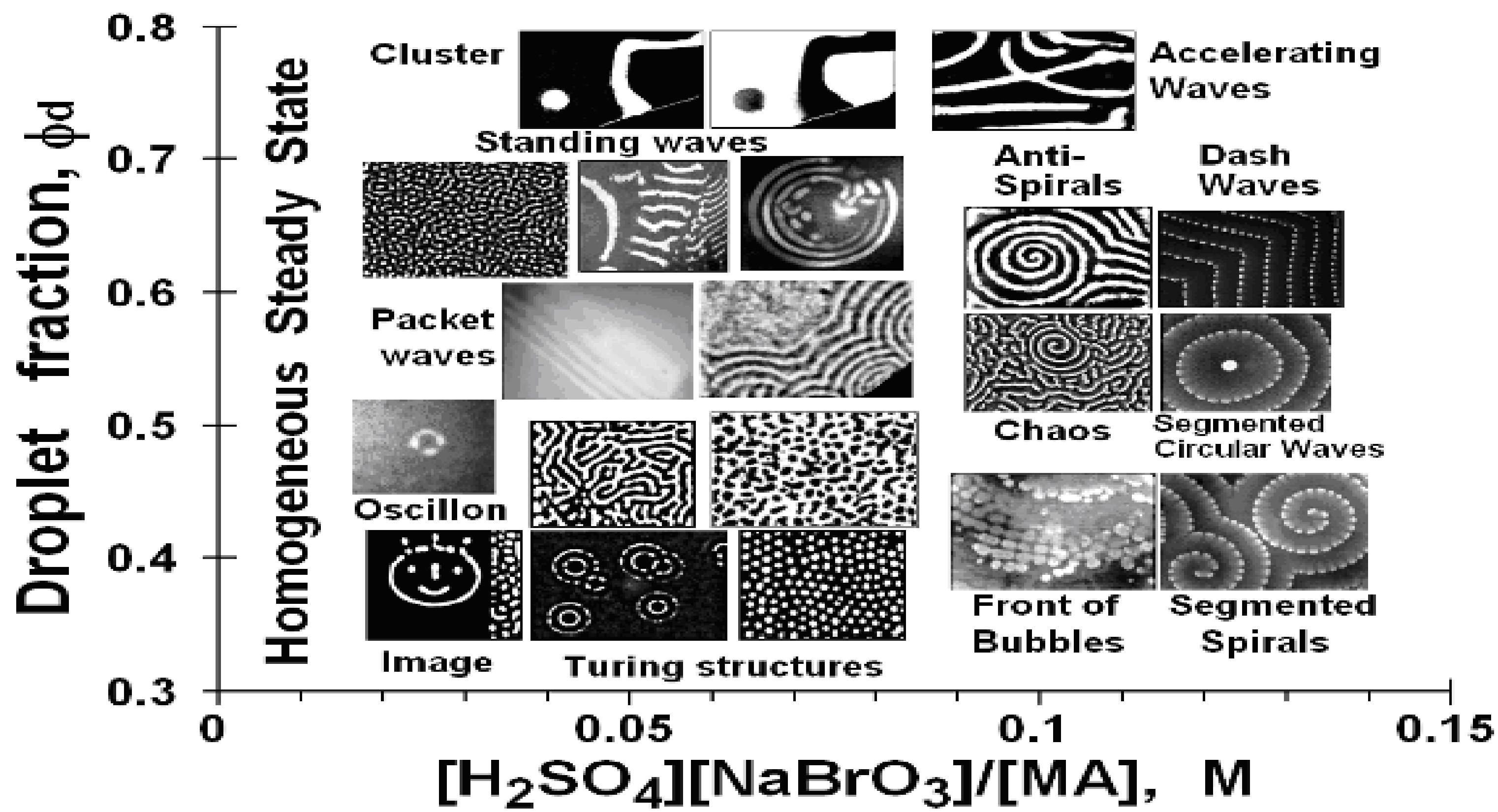




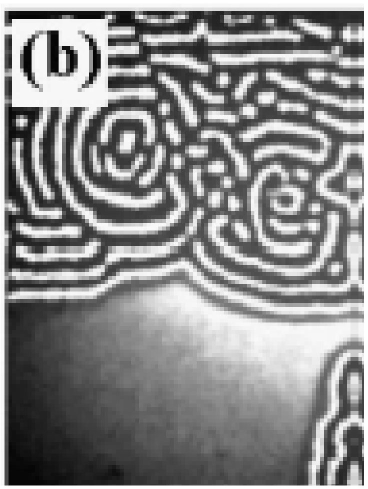
Two flat optical windows 50 mm in diameter are separated by an annular Teflon gasket with inner and outer diameters 20 and 47 mm, respectively, and a thickness of 0.1 mm. The reaction volume (red) is thus a closed cylinder of radius 10 mm and height 0.1 mm.

Patterns are observed through a microscope equipped with a digital CCD camera connected to a personal computer.

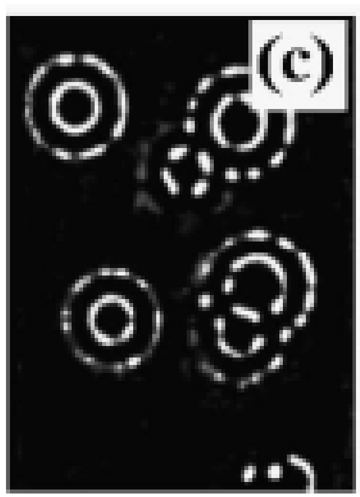
BZ-AOT PATTERNS



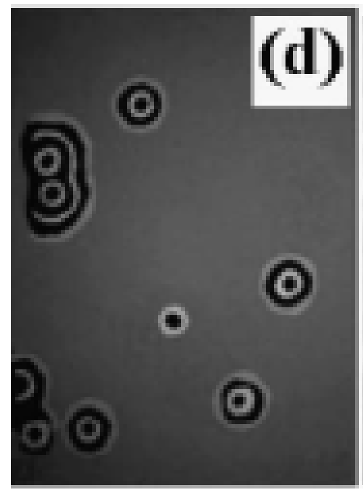
Localized patterns in BZ-AOT



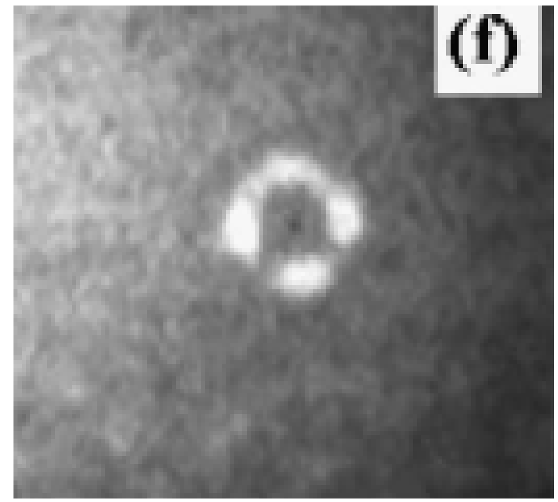
Ferroin, Turing



Ru(bipy), Turing



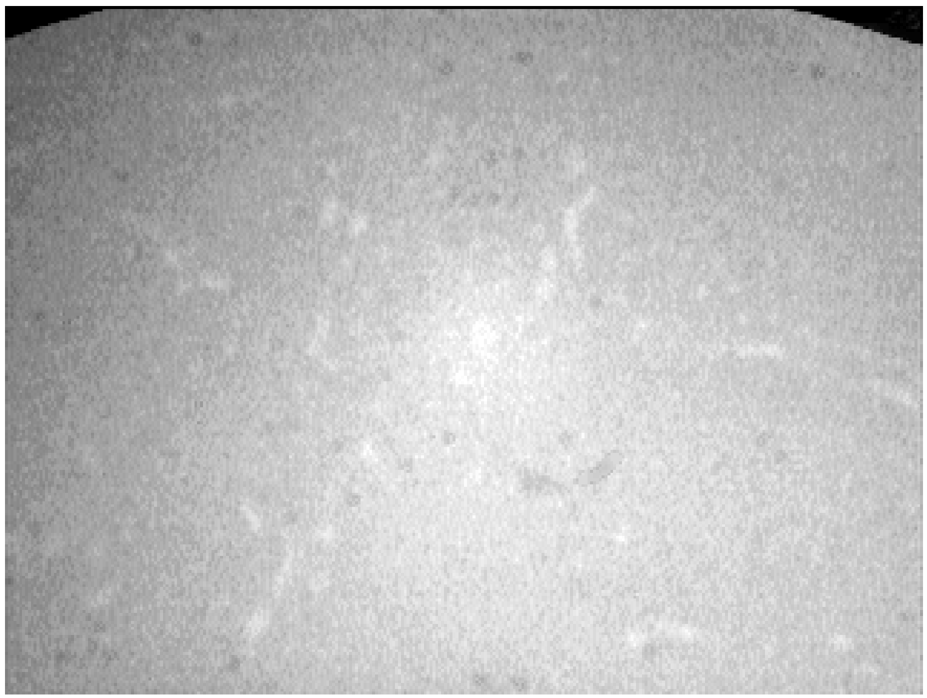
Ferroin, Oscillon



Ru(bipy), Oscillon

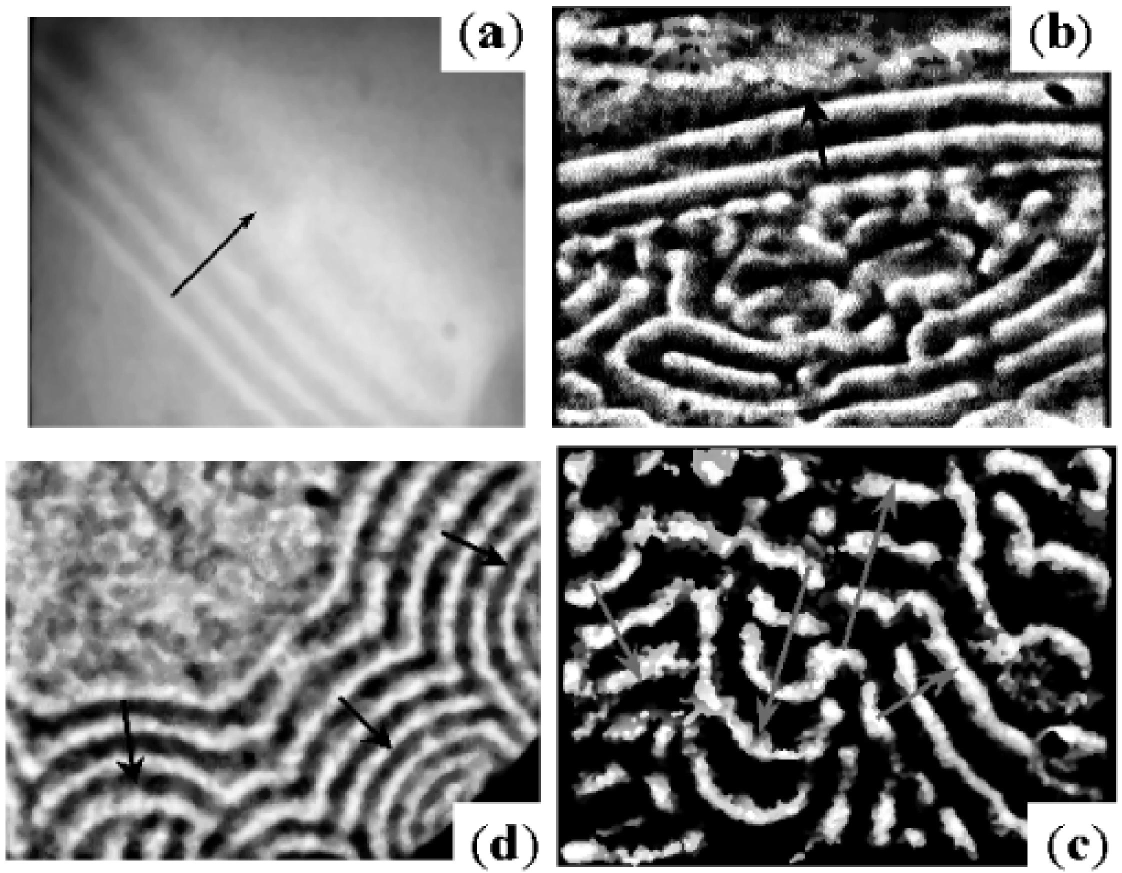
Vanag & Epstein, PRL 92, 128301 (2004)

Localized oscillons



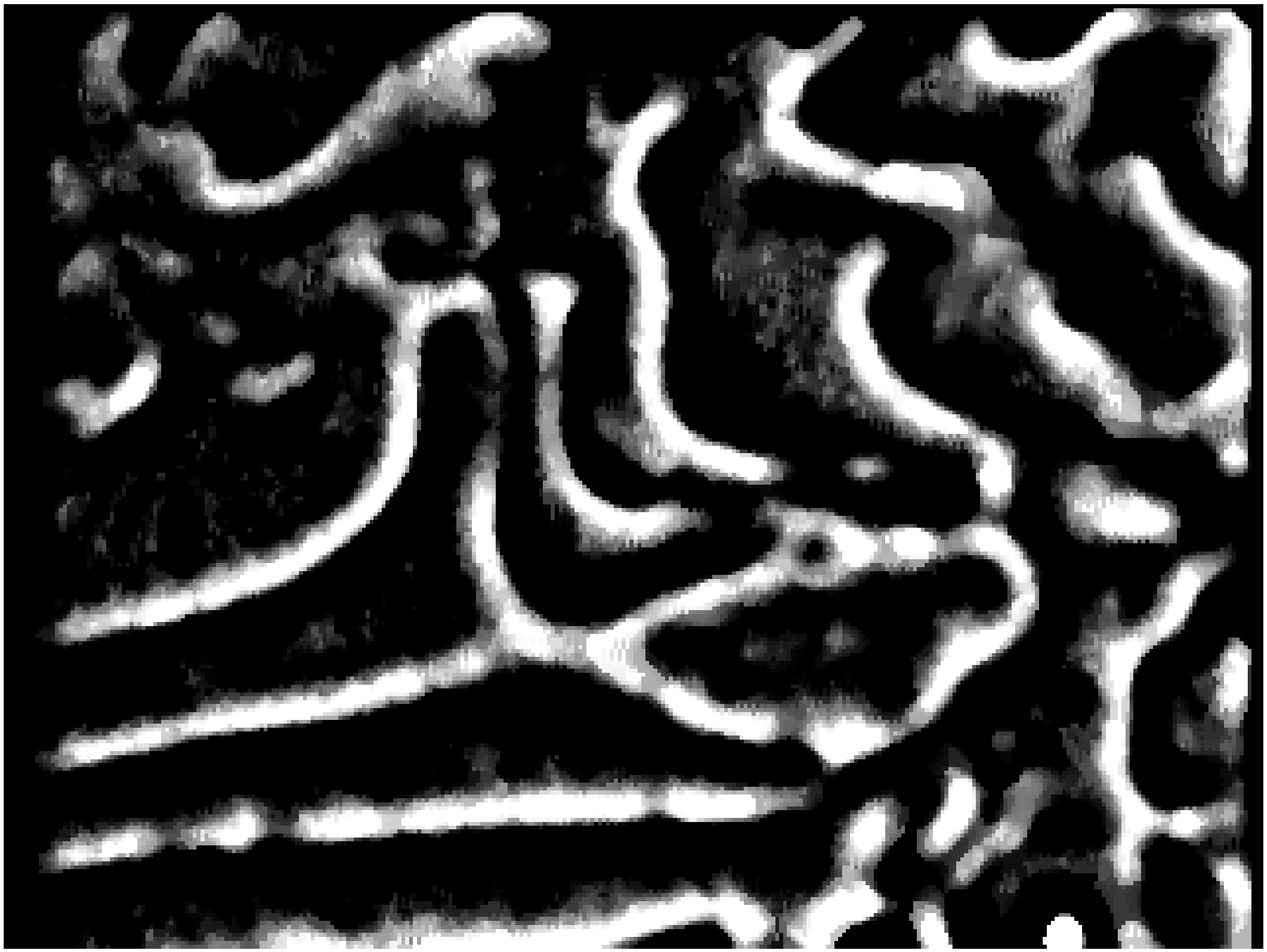
Vanag & Epstein, PRL 92, 128301 (2004

Packet Waves
(A dynamic analog of snaking??)

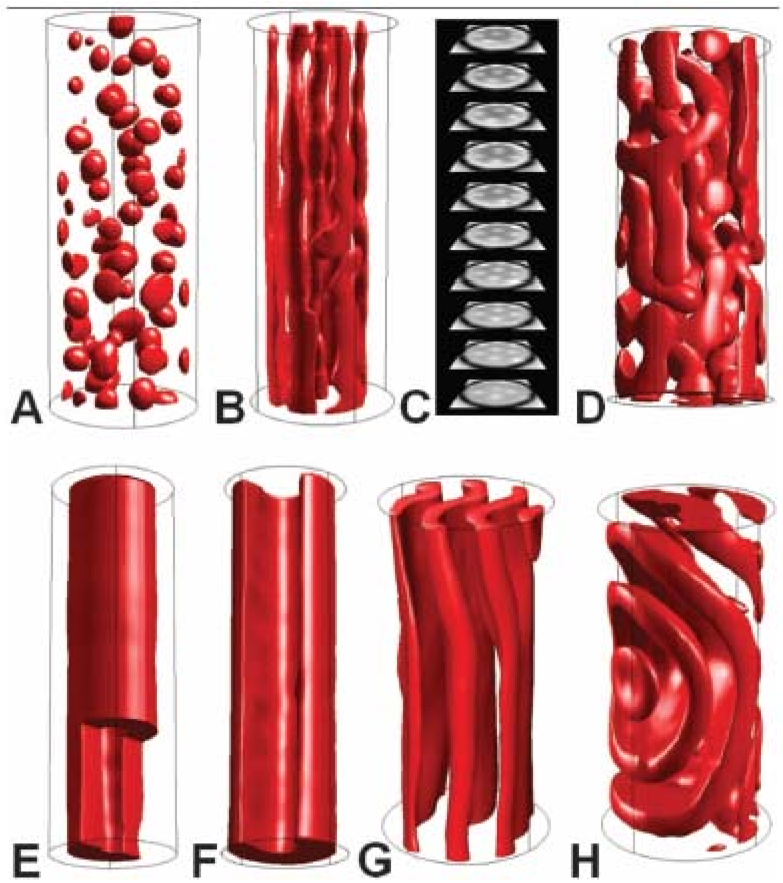


Vanag and Epstein, PRL 88, 088303 (2002)

X-waves (?)



Tomographically reconstructed 3D Turing patterns



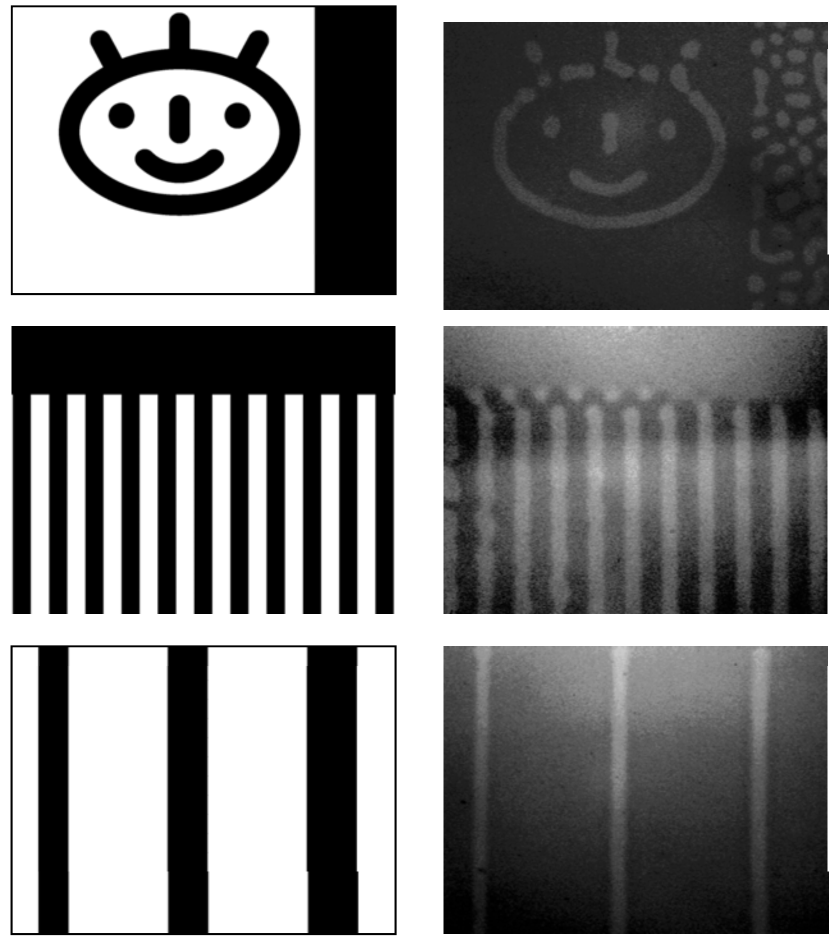
Bansagi, Vanag & Epstein, Science 331, 1309 (2011)

Simulations of 3D localized spots – Leda, Vanag & Epstein, PRE 80, 066204 (2009)

Subcritical bifurcation – BZ-AOT

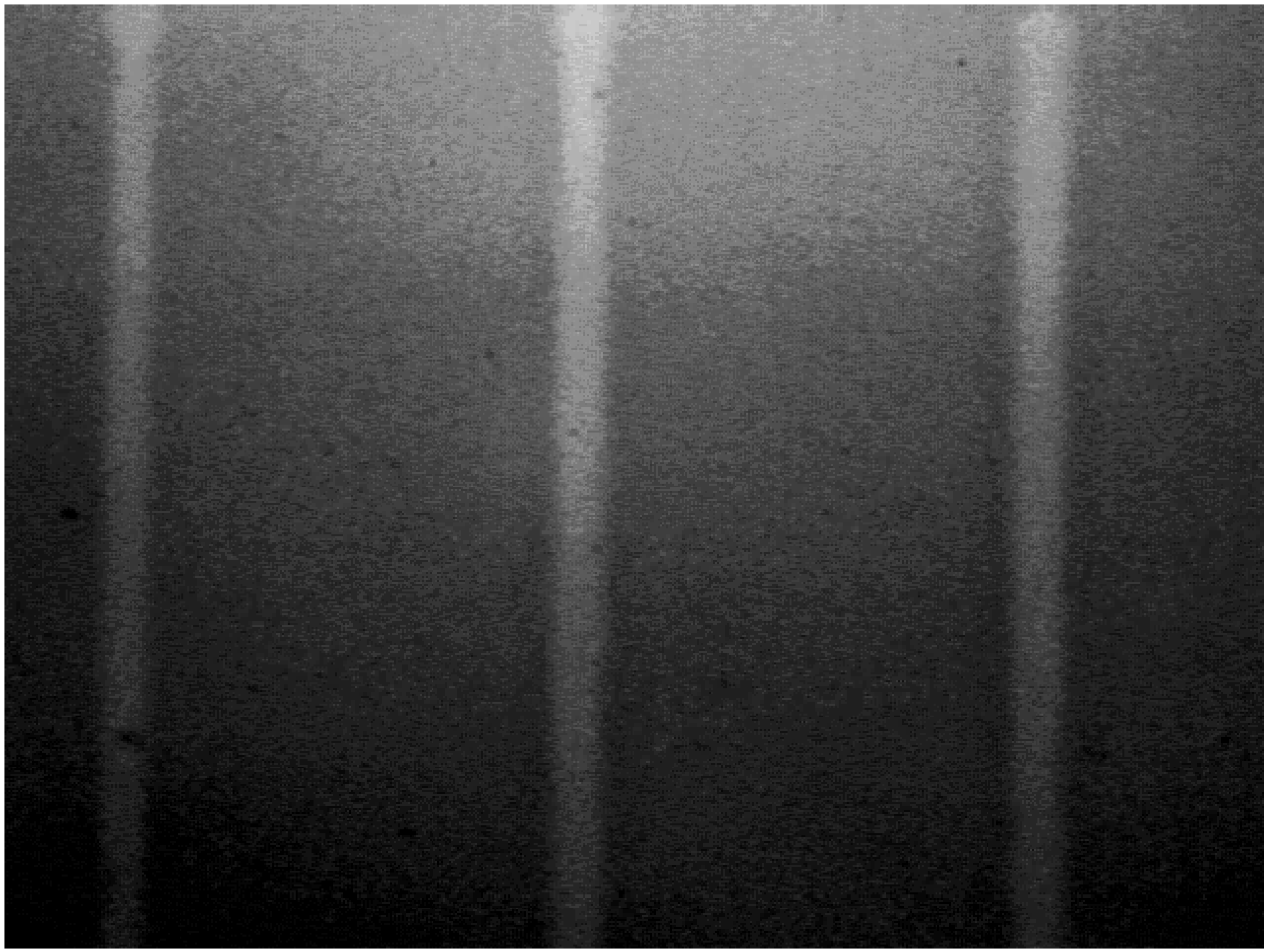
1. Maximum I to erase all patterns.
2. Insert mask.
3. Lower I into bistable range.
4. Remove mask.

Patterns disappear and then return if I is raised briefly above bistable range.

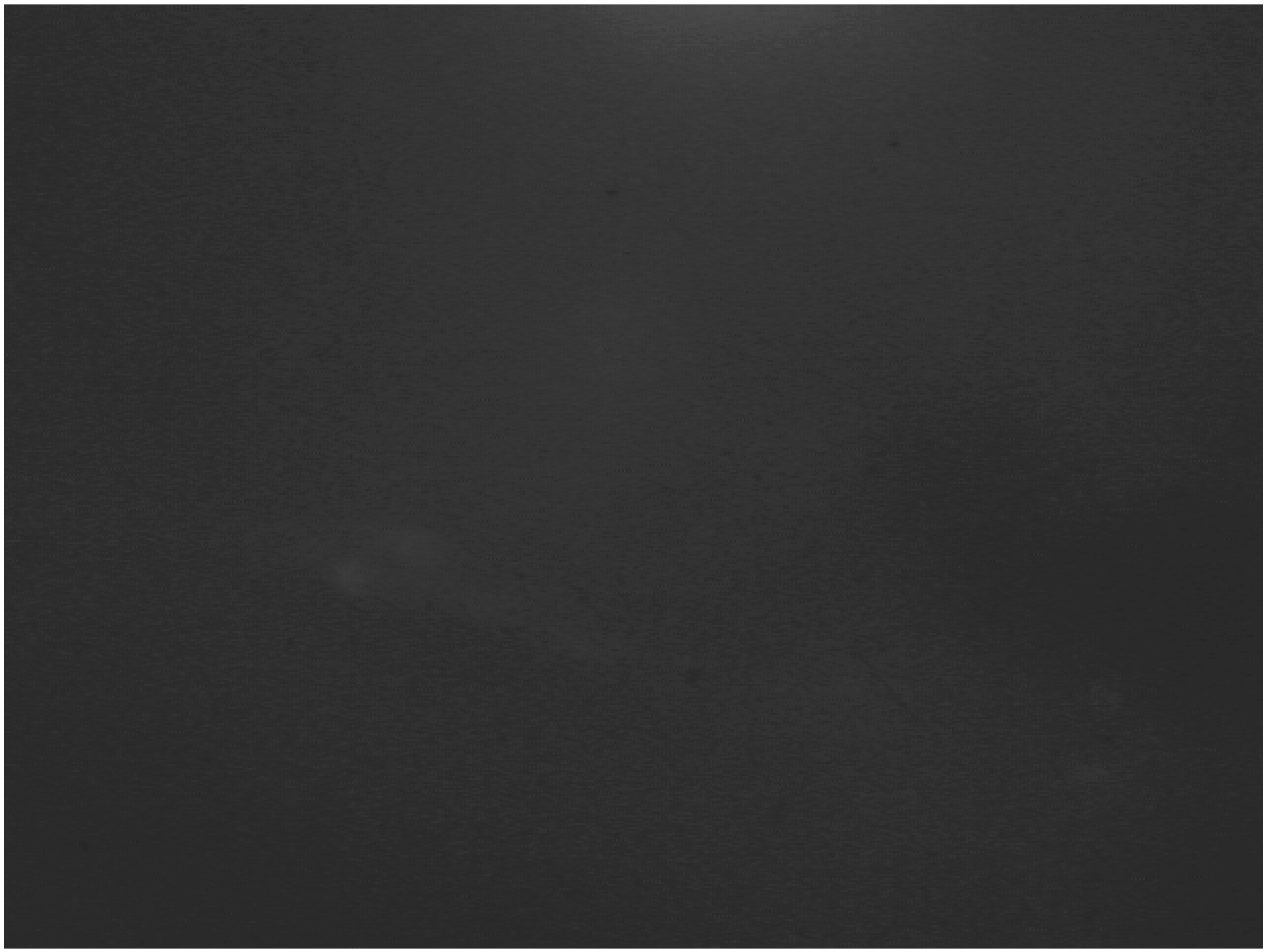


Kaminaga, Vanag & Epstein, Angew. Chem. Int. Ed. 45, 3087 (2006)

BZ-AOT localized waves



BZ-AOT memory



Are localized structures good for anything?

CHAOS VOLUME 14, NUMBER 1 MARCH 2004

A new approach to data storage using localized structures

P. Coullet[a)]
INLN, 1361 Route des Lucioles, 06560 Valbonne, France

C. Riera[b)]
DAMTP, Silver Street, Cambridge CB3 9EW, United Kingdom

C. Tresser[c)]
IBM, P.O. Box 218, Yorktown Heights, New York 10598

(Received 22 April 2003; accepted 1 December 2003; published online 9 February 2004)

In this paper we describe how to use the bifurcation structure of static localized solutions in one dimension to store information on a medium in such a way that no extrinsic grid is needed to locate the information. We demonstrate that these principles, deduced from the mathematics adapted to describe one-dimensional media, also allow one to store information on two-dimensional media.
 [DOI: 10.1063/1.1642311]

Could you build a computer (or a brain)? Need memory <u>and</u> processing.

Modeling

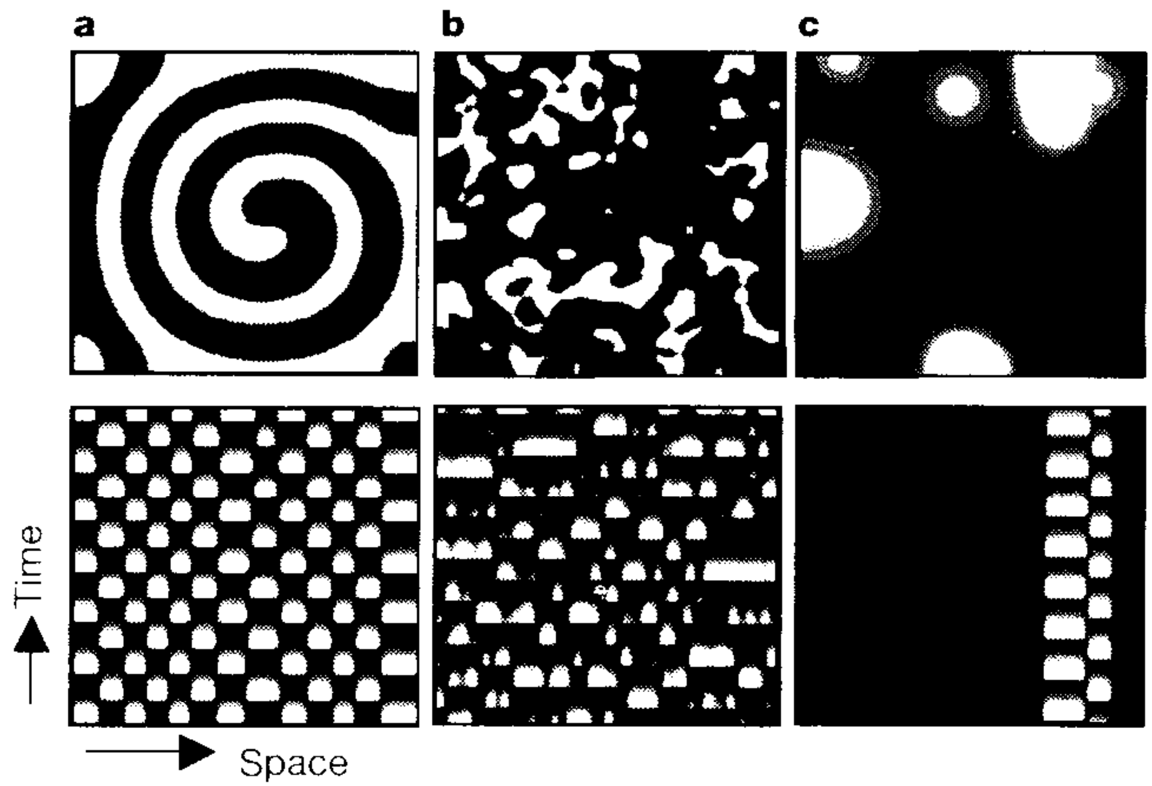
- Kruskal – solitons
- Koga and Kuramoto – reaction-diffusion (FHN)
- Kerner & Osipov – dissipative solitons
- Krischer & Mikhailov – bifurcation to traveling spots (surface catalysis)
- Ataullakhonov – blood clotting model
- Purwins – reaction-diffusion models

Add new variables

$Y \to X$	k1	$X = HBrO_2$	
$Y + X \to 0$	k2	$Y = Br^-$	
$X \to 2X + 2Z$	k3	Z = ferriin	Oregonator
$2X \to 0$	k4		
$Z \to hY$		k5	
$X \to S$	kf	$S = Br_2O_4$ (or $BrO_2^\bullet$)	
$S \to X$	kb	in the oil phase	
$Y \to V$	kf'	$V = Br_2$ in the oil phase	
$V \to Y$	kb'		

$D_V, D_S \gg D_X, D_Y, D_Z$

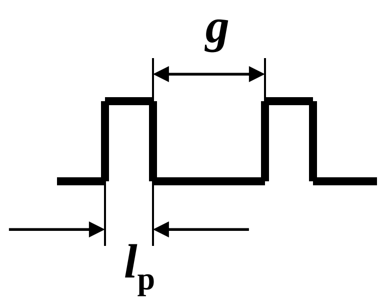
Simulations – Global feedback

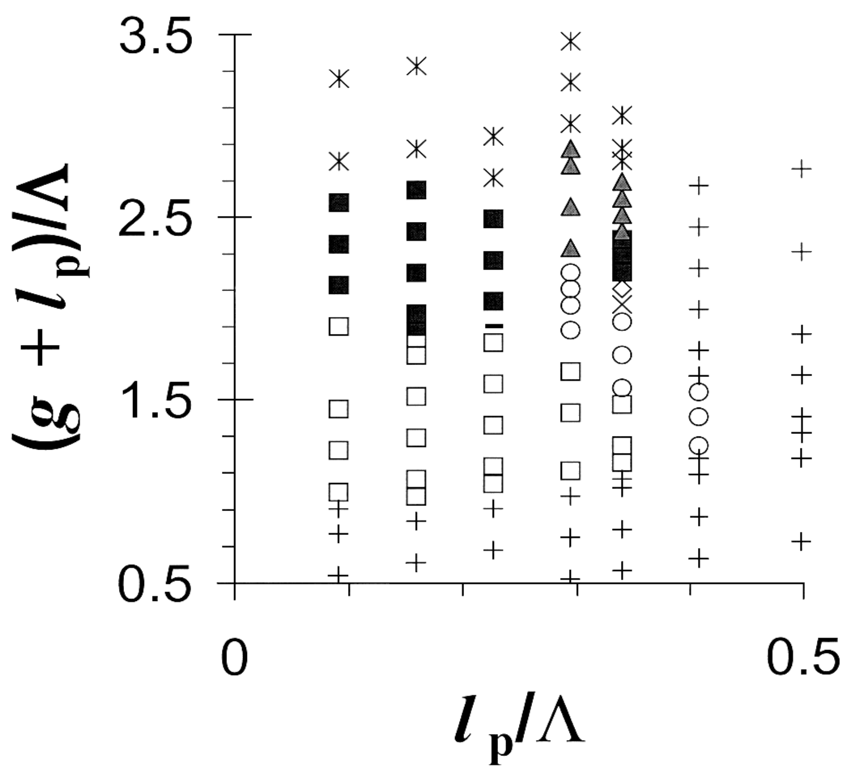


Vanag,Yang, Dolnik, Zhabotinsky & Epstein, Nature 406, 389 (2000)

Interaction of Localized Peaks

Initial perturbation:





Two identical tooth-like initial perturbations are separated by a gap of length g. +, SS; □, oscillon with two synchronously oscillating peaks; ○, stationary Turing pattern with two peaks; ■, oscillon with three synchronously oscillating peaks; ▲, pattern with three peaks, the middle one oscillating and the outer ones stationary (T+O+T); ×, single oscillon; −, single stationary peak; ◊, oscillon with two peaks oscillating anti-phase; ∗, two independent Turing or oscillatory peaks.

Vanag & Epstein, PRL 92, 128301 (2004)

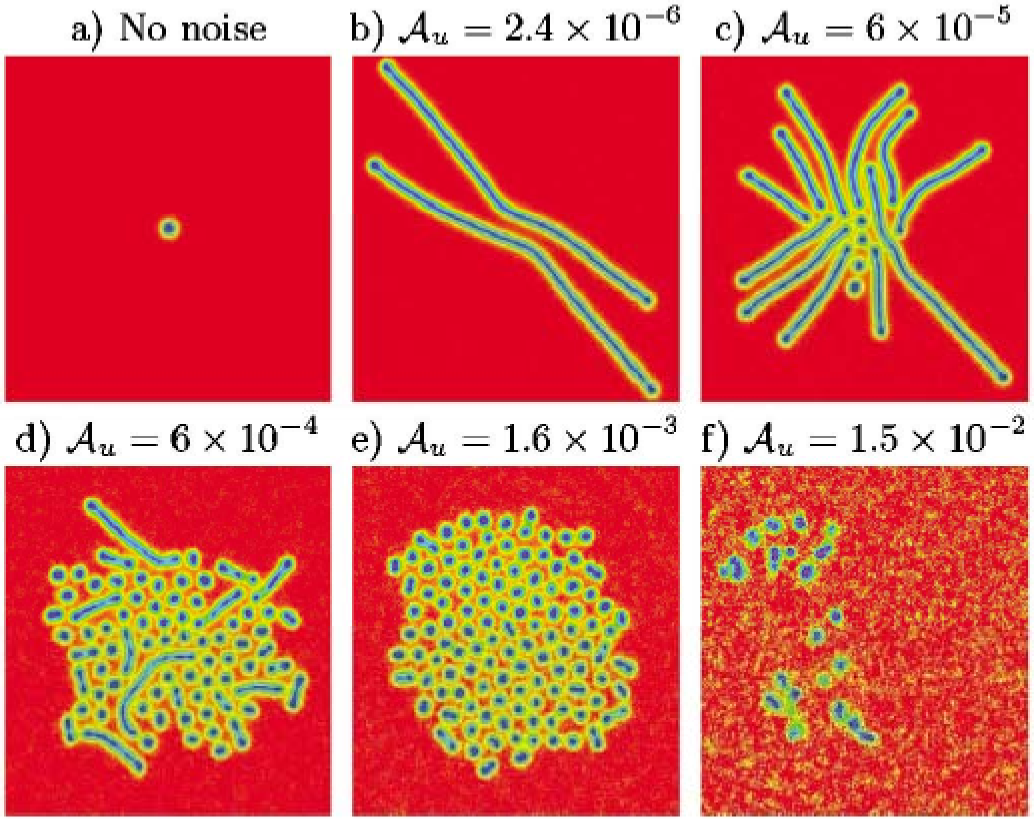
Some further questions

- What are the bifurcation scenarios for transitions between localized patterns and between localized and global patterns?
- What can be said about the spatial extent, shape, and amplitude of perturbations needed to initiate a particular type of localized pattern?
- How do localized spots interact with one another?
- What kinds of complex patterns can be built from individual localized spots?

Conclusions

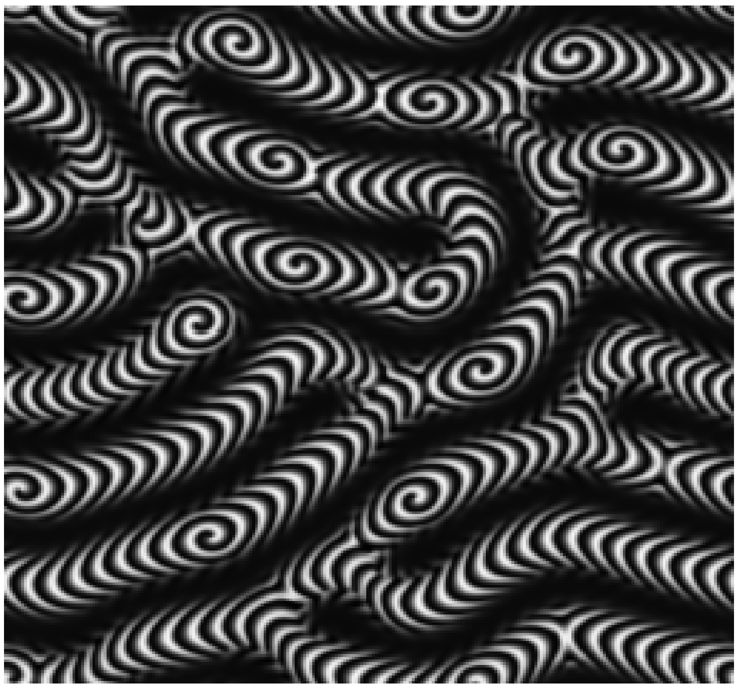
- A rich variety of localized structures can be generated in reaction-diffusion systems by several techniques, including global negative feedback, periodic perturbation and microemulsions.
- They may provide an attractive vehicle for information storage.
- They can be numerically simulated with chemically plausible models, but much room remains to develop a good theoretical understanding.

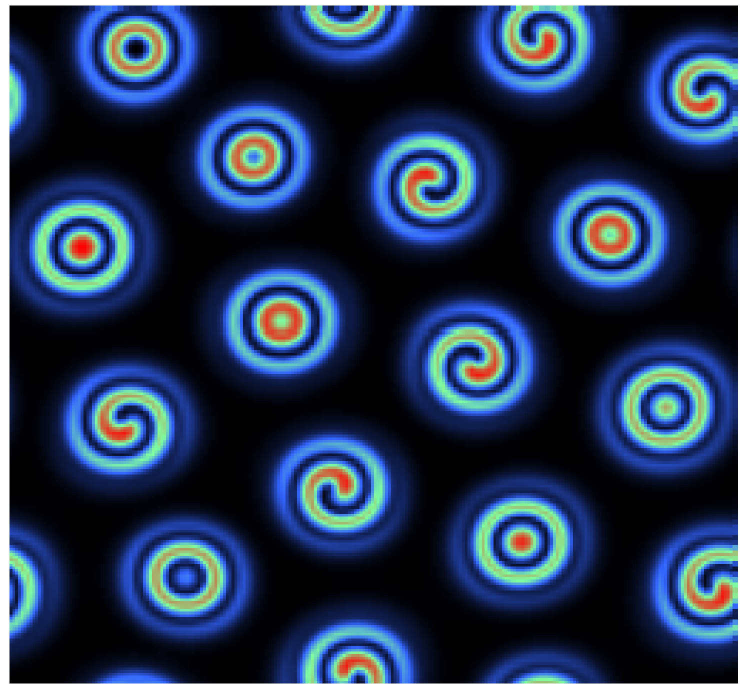
Noise induced localized patterns in the Gray-Scott model model



Lesmes, Hochberg, Moran & Perez-Mercader, PRL 91, 238301 (2003)

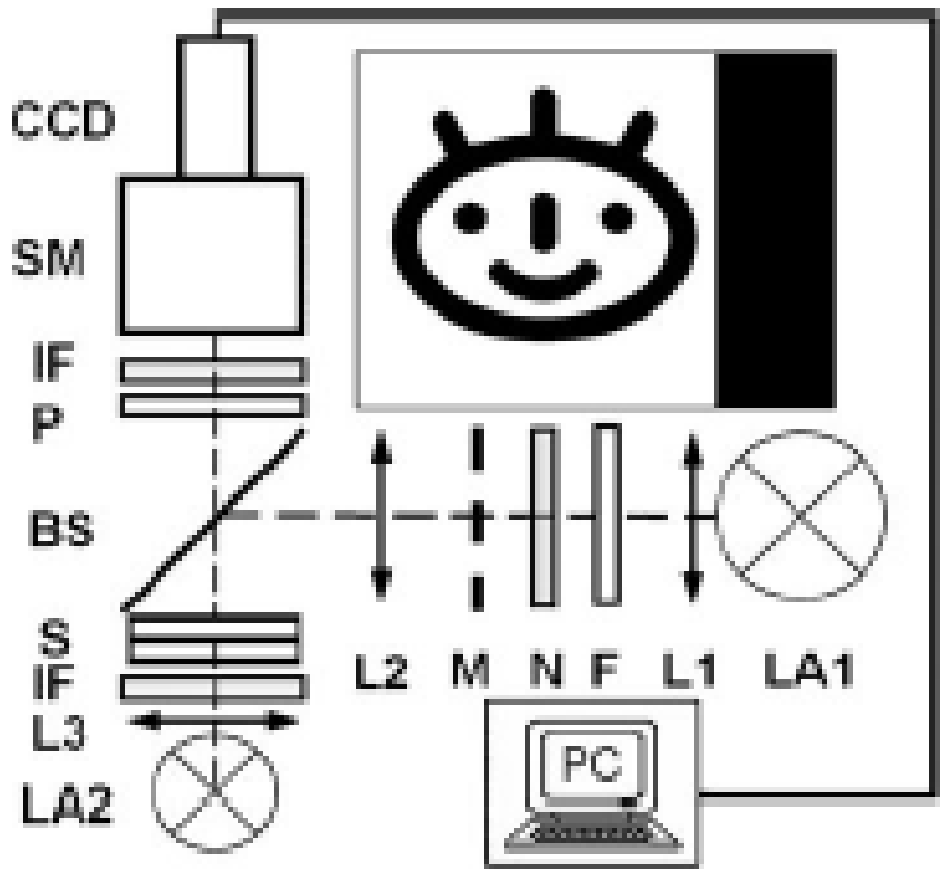
Coupled layer model – Patterns within patterns



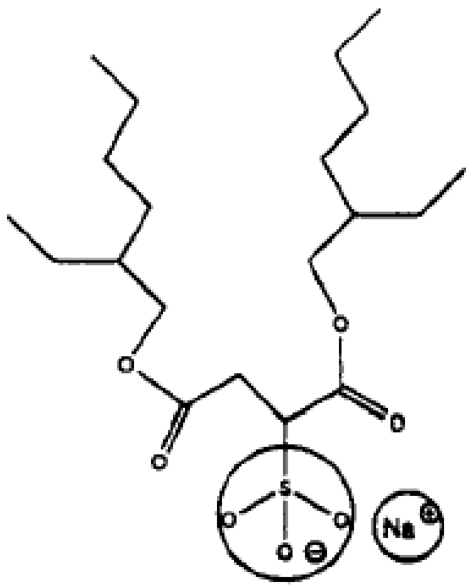


Yang & Epstein, *Phys. Rev. Lett*. **90**, 178303 (2003).

Subcritical bifurcation – BZ-AOT

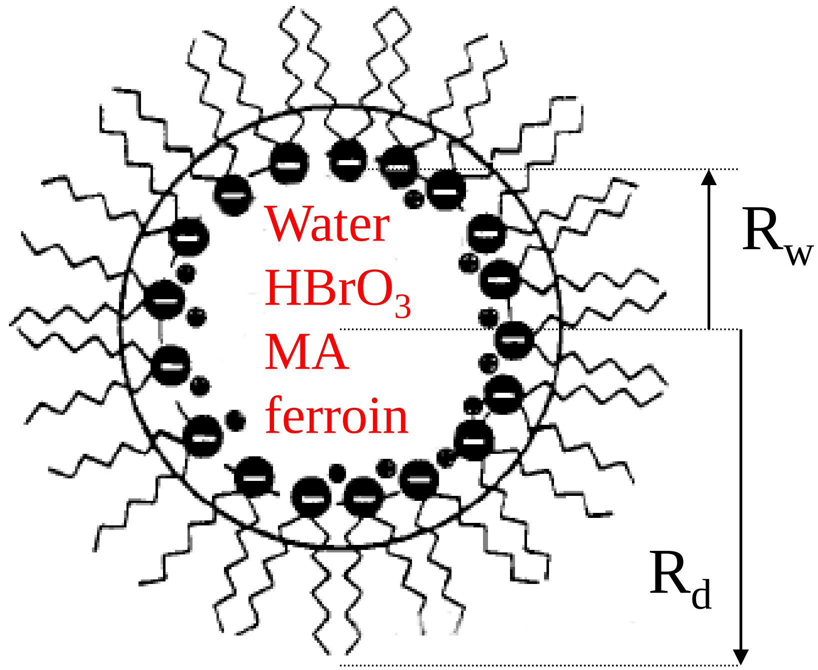


AOT reverse micelles or water-in-oil microemulsion



Aerosol-OT

(sodium bis(2-ethylhexyl) sulfosuccinate)



Oil = $CH_3(CH_2)_nCH_3$

BZ reactants reside in the micelle water cores

MA = malonic acid

R_w = radius of water core

R_d = radius of a droplet,

$$\omega = \frac{[H_2O]}{[AOT]}$$

$R_w = 0.17\omega$

R_d = 3 - 4 nm

ϕ_d = volume fraction of dispersed phase (water plus surfactant)

Modeling – add third variable

$\partial x/\partial t = k_1 - k_2x - k_4x + k_5yx^4/(K^4 + x^4) + k_6y$ $\mathbf{- k_7x + k_8z}$ $+ D_x\nabla^2x$

$\partial y/\partial t = k_4x - k_5yx^4/(K^4 + x^4) - k_6y + D_y\nabla^2y$

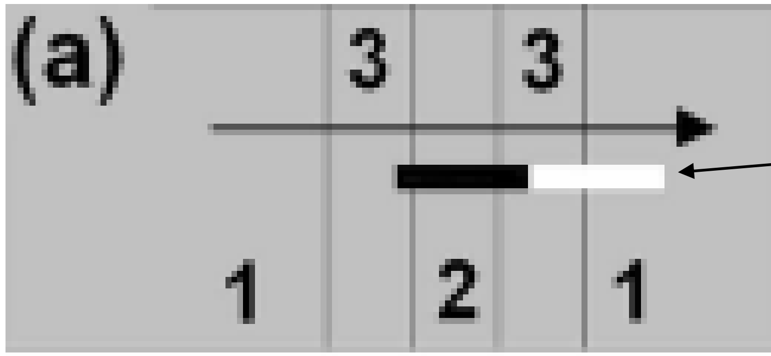
$\mathbf{\partial z/\partial t = k_7x - k_8z + D_z\nabla^2z}$

Black = Samogyi-Stucki model for calcium oscillations

Red = added terms to generate localized oscillons

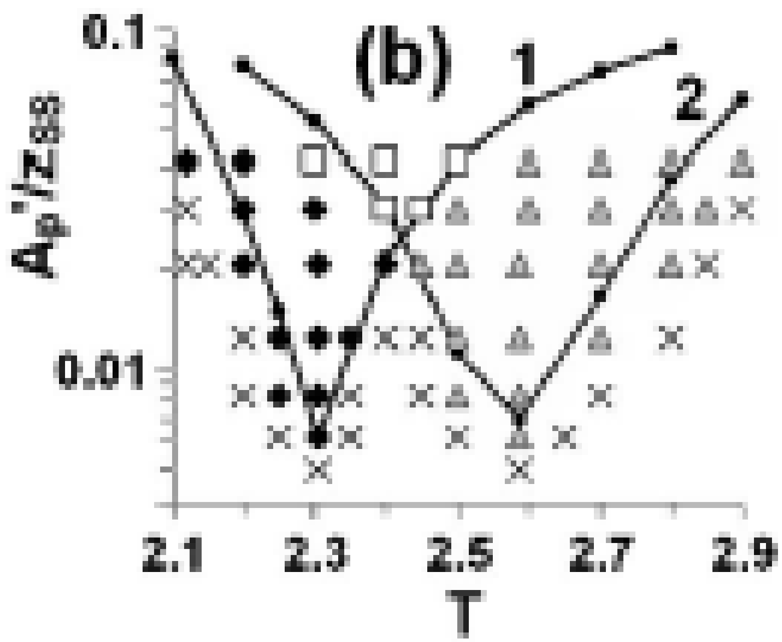
Resonance-Induced Oscillons

1&2 support oscillon
3 steady state only



Traveling perturbation

Can filter mixed signals:
$\Sigma A_{pi'} \sin(2\pi t/T_{pi}) \varphi(y)$



Vanag & Epstein, PRE 73, 016201 (2006)